Optical Frequency Standards and Measurements

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Abstract ----We describe the performance characteristics and frequency measurements of two high-accuracy high-stability laser-cooled atomic frequency standards. One is a 657 nm (456 THz) reference using magneto-optically trapped Ca atoms, and the other is a 282 nm (1 064 THz) reference based on a single Hg^+ ion confined in an rf-Paul trap. A femtosecond mode-locked laser combined with a nonlinear microstructure fiber produces a broad and stable comb of optical modes that is used to measure the frequencies of the reference lasers locked to the atomic standards. The measurement system is referenced to the primary frequency standard, NIST F-1, a Cs atomic fountain clock. Both optical standards demonstrate exceptional short-term instability (≈ $5x10^{-15}$ at 1 second) as well as excellent reproducibility over time. In light of our expectations for the future of optical frequency standards, we consider the present performance of the femtosecond optical frequency comb, along with its limitations and future requirements.

I. INTRODUCTION

Spectrally narrow optical transitions in atoms and ions probed by stable lasers are now emerging as the next generation of high-accuracy, high-stability frequency standards. The principle advantage of optical standards over their well-known microwave counterparts is the higher operating frequency; this opens the potential for orders of magnitude better frequency stability, in principle, by the ratio of the operating frequencies $f_{optical}/f_{microwave} \approx 10^5$. With innovative developments of the past few years, including very stable lasers and a new practical method for counting optical frequencies, it appears that we now have the tools to realize the potential of optical frequency references.

We focus here on two different optical frequency standards: one is a 657 nm (456 THz) standard using $\approx 10^7$ laser-cooled Ca atoms, and the other, a 282 nm (1,064 THz) standard using a single trapped and laser-cooled Hg^+ ion. After briefly describing the operation of these standards we outline their present performance and provide some glimpse of future potential. With simplifying assumptions we estimate the stability that could be achieved with cold atom optical frequency references. We then describe our femtosecond (fs) mode-locked laser-based optical frequency measuring system that uses the recent concepts and developments from H≅nsch and collaborators, and Hall and collaborators [1]. This system was recently used to measure the frequency of the two optical standards relative to the Cs primary frequency standard at NIST. Questions of absolute accuracy of the optical standards will require detailed evaluation of systematic errors and uncertainties that can only be done with the comparison of multiple standards. This will have to wait for the future, but the femtosecond optical combs now provide a convenient means to make direct intercomparisons between different optical standards as well as a coherent connection between the RF and optical domains. Due in large part to this new measurement capability, research groups around the world are pursuing optical frequency standards with renewed enthusiasm.

We also consider some fundamental and practical limits to the performance of our femtosecond optical frequency comb. The stability and projected accuracy of the optical standards put stringent requirements on the performance of the optical frequency metrology. Nonetheless, these systems already allow us to explore a regime of atomic frequency stability that is well beyond what has been possible with microwave atomic standards.

The general goal of this paper is to provide a status report on the rapidly changing state of our field. At this point, we can safely predict that the present results will soon be outdated. This is truly a revolutionary time for atomic frequency standards and measurements. We are just beginning to experience the tremendous advances in stability and accuracy that were previously imagined, but which were not possible to explore until now.

The rapid improvements are reminiscent of the jump in accuracy that occurred with the pioneering work of Jennings et al. in 1983 when they first demonstrated that optical frequencies could be measured using a harmonic frequency chain. Their system consisted of a series of lasers with ever increasing frequencies locked in a sequence of harmonics from the microwaves up to the visible part of the spectrum [2,3]. From that moment until the late '90s, the precision in optical frequency measurements continued to improve, in terms of fractional frequency, from ≈ 10^{-10} to 10^{-13}. Now, the combined effects of better stabilized lasers, better atomic references using cold atoms and ions, and fs-optical-frequency-metrology, have advanced the performance by almost two orders of magnitude in just 2 years. Moreover, the femtosecond mode-locked laser-based measuring systems are also more practical to use. Historically, new optical frequency measurements had been

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reported every few years; but in the brief time since the introduction in 1999 of octave-spanning optical-frequency combs, more than 10 new optical frequency measurements have been reported.

II. Laser-cooled atoms and ions as optical Atomic frequency references

We are presently developing two optical frequency standards. Both have been described in detail previously [4,5] so we include only a brief description here.

A. Ion considerations

Trapped ions, particularly single laser-cooled ions, have numerous advantages as optical frequency standards and clocks [6,7]. Most importantly, ions can be confined in an RF trap and laser-cooled such that the amplitude of the residual motion is much less than the optical wavelength of the probe radiation (the so-called Lamb-Dicke limit). This nearly eliminates the velocity-dependent Doppler broadening and shifts associated with motion of the ion relative to the probing radiation. In a cryogenic environment the ion is nearly unperturbed by atomic collisions, and the effects of blackbody radiation are also very small. The storage time of a single ion in a trap can be months; hence, the probe interaction time is not constrained, which permits extremely high-Q resonances to be observed. All of these factors are critically important if we hope to achieve the highest accuracy.

The technical challenges of making an optical frequency standard based on a single ion are formidable; but single-ion standards have now been achieved in a handful of laboratories around the world [8,9]. At NIST we are developing an optical frequency standard based on a single trapped $^{199}Hg^+$ ion. The performance of this standard is immediately competitive with the performance of the best microwave standards and has the potential to surpass those standards in terms of stability, frequency reproducibility, and accuracy.

A single $^{199}Hg^+$ ion is trapped in a small RF Paul trap (≈ 1 mm internal dimensions) and laser-cooled to a few millikelvins using 194 nm radiation. The relevant energy levels for the cooling and clock transitions are shown in fig. 1. A highly stabilized dye laser at 563 nm with a linewidth of less than 1 Hz is frequency doubled to 282 nm (1,064 THz) to probe the clock transition [10]. Measured linewidths

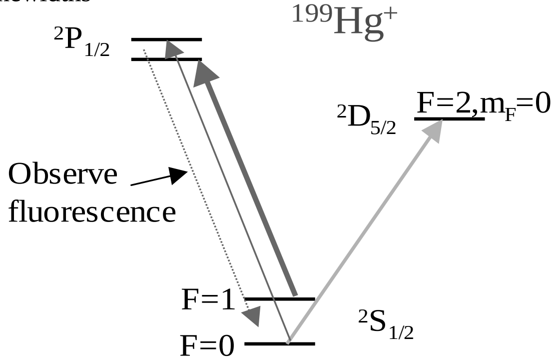

Fig. 1. Hg^+ energy-level diagram.

as narrow as 6.7 Hz on the 282 nm transition have recently been reported [11]. For an averaging time τ in seconds, the projected instability of an optical frequency standard using a single Hg^+ ion is $<1x10^{-15}\tau^{-1/2}$ and fractional frequency uncertainties approaching $1x10^{-18}$ seem feasible [12].

B. Neutral atom considerations

Some neutral atoms also have narrow optical transitions that are relatively insensitive to external perturbations and are thus attractive as optical frequency standards [13]. Neutral atoms have some advantages and disadvantages relative to ions. Using the well-established techniques of laser cooling and trapping, they are fairly easy to confine and cool to low temperatures. However, in contrast to ions, the trapping methods for neutrals perturb the atomic energy levels, which is unacceptable for use in a frequency standard. To avoid the broadening and shifts associated with the trap, neutral atoms are released from the trap before the clock transition is probed. The atoms fall from the trap under the influence of gravity and expand with low thermal velocities (typically a few cm/s). The resulting atomic motion brings with it serious limitations in accuracy (and even stability) that are associated with velocity dependent frequency shifts. Two of the more troublesome effects are the limited observation time, and the incomplete cancellation of the first order Doppler shift associated with wave-front curvature and k-vector mismatch. These motional effects pose serious limitations to the ultimate accuracy that might be achieved with neutral atoms in a gravitational potential. Reduced observation times limit the line-Q, the stability and the accuracy. However, neutral atoms do have at least one significant advantage, that is that large numbers of atoms can be used, producing a large signal-to-noise ratio in a short time, and the potential for exceptional short-term stability.

The atomic Ca optical frequency standard is one of the promising cases, because it has narrow atomic resonances that are reasonably immune from external perturbations, it is readily laser-cooled and trapped, and it is experimentally convenient because the relevant transitions are accessible with tunable diode lasers.[5] A simplified energy-level diagram is shown in fig.2.

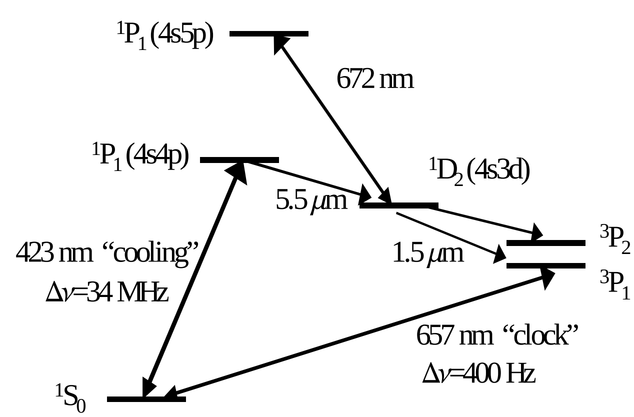

Fig. 2. Ca energy-level diagram.

A frequency doubled diode laser tuned to the 423 nm transition cools and traps about 10^7 Ca atoms in a MOT in about 5 ms. The cooling radiation is turned off, and an injection-locked and stabilized diode laser at 657 nm (456 THz) probes the clock transition with the separated excitation method of optical Ramsey fringes [14,15]. The actual excitation technique we employ is the 4-pulse traveling wave method introduced by Bordϑ [16,17]. Optical fringes with high signal-to-noise ratio are observed using shelving detection on the cooling transition. The measured fringe widths depend on the Ramsey time of the probe field and range from 200 Hz to 11.5 kHz. The present Ca standard can provide short-term fractional frequency instability of about $4\times10^{-15}\tau^{-1/2}$ using 960 Hz wide fringes [18]. With modifications to the apparatus and lower temperature Ca atoms, we estimate that this system should reach instabilities of about $1\times10^{-16}\tau^{-1/2}$. Until recently, we have not focused much attention on controlling, or evaluating, the systematic frequency shifts in our Ca standard. With the advent of femtosecond-optical-frequency-metrology (discussed below) and the close proximity (180 m of optical fiber distance) of the Hg^+ standard and the Cs fountain standard, we can now make high-accuracy inter-comparisons and begin to study the systematic effects that will ultimately determine the accuracy. We are just beginning this evaluation process, and as might be anticipated the velocity-dependent frequency shifts appear to be the most challenging to control. With our present experimental apparatus, which has not been optimized for accuracy, we assign an uncertainty to our Ca optical frequency standard of +/-26 Hz at 456 THz [19].

III. STABILITY REQUIREMENTS AND POTENTIAL

Fractional frequency uncertainties as small as 1×10^{-18} have been predicted for single trapped-ion optical standards [12]. This ambitious goal is three orders of magnitude beyond the present state of the art frequency standards and brings to focus many technical challenges that will need to be addressed. For the next generation of atomic clocks to reach this level of accuracy will require exceptional short-term stability.

We can estimate the short-term stability that is achievable with atomic standards under the simplifying assumptions that the frequency noise of the local oscillator (in this case the stabilized laser) can be neglected [20], and that we can achieve atom projection-noise-limited detection[21,22]. The stability, or rather instability, is most commonly expressed as the two-sample Allan deviation, which, as outlined in appendix A gives the fractional frequency instability as a function of the averaging time τ. We assume an atomic resonance centered at frequency ν_0 with linewidth $\Delta\nu$ (FWHM), and that the system uses the Ramsey separated-fields method with Ramsey time T_R, and detects N_o atoms on one side of the atomic resonance. A full measurement of both sides of the atomic resonance line determines the line center, and is completed in a cycle time T_c.

Under these assumptions and the analysis of appendix A, the fractional frequency instability for an atomic frequency reference (given by the Allan Deviation) can be written as,

$$\sigma_y(\tau) = \frac{\delta\nu(\tau)_{rms}}{\nu_0} = \frac{\Delta\nu_{Ram}}{\pi\nu_0}\sqrt{\frac{T_C}{2N_0\tau}} \quad .$$

This expression assumes a sinusoidal fringe shape with 100 % contrast, a fringe-width given in terms of the Ramsey time T_R, the optimum transition probability at resonance $p_0 = 1$, and the optimum detuning $\delta_0=(\Delta\nu_{ram})/2$ for maximum slope. In most real experiments these ideal conditions are not satisfied and so the instability is greater than that predicted. Thus, for a sinusoidal fringe shape but with reduced contrast $0 \le p_0 \le 1$, a FWHM linewidth $\Delta\nu$, and a nominal detuning from resonance δ_0, we define a lineshape slope factor C,

$$C = p_0 sin[\pi\delta_0 / \Delta\nu] \quad .$$

This leads to the following form for the instability,

$$\sigma_y(\tau) = \frac{\Delta\nu}{C\pi\nu_0}\sqrt{\frac{p_+(1-p_+)+p_-(1-p_-)}{N_0}}\sqrt{\frac{T_C}{\tau}} \,.$$

Here p_+ and p_- are the transition probabilities for the two sides of the line at nominal detuning from resonance $\pm\,\delta_0$.

If the cycle time T_c is significantly larger than $1/\Delta\nu$, then there is excess dead time in the measurement cycle and the stability is degraded. This is often the case in present day standards because of the time required used for laser cooling and trapping, or waiting for an excited state to decay, or because of long detection times dictated by poor signal to noise.

In the projection noise limit, $\sigma_y(\tau)$ varies as the inverse square root of the averaging time τ, which means that to reach high accuracy in a reasonable time we require excellent short-term stability. For example, high-quality quartz-based local oscillators have instabilities of about 1×10^{-13}, and thus microwave atomic fountain clocks have instabilities of about $1\times10^{-13}\tau^{-1/2}$. With this instability it requires about 3 hours of averaging to reach their present uncertainty limit at $\approx1\times10^{-15}$. Our optical standards can reach $\approx1\times10^{-15}$ instability in about ten seconds and have the potential to go well beyond this. (Methods to improve the short-term instability of microwave atomic standards are also being pursued [23], but from the perspective of an outsider it appears to be an uphill battle compared to what is already possible using optical frequencies.) Fig. 3 shows the short-term instability of some of high stability frequency references.

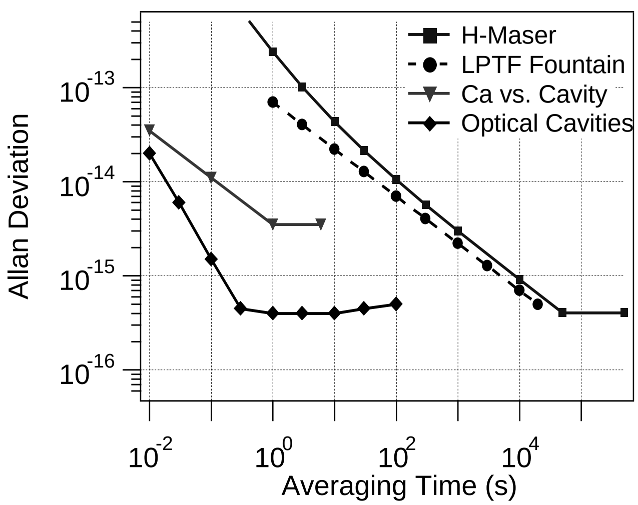

Fig. 3. Measured frequency instabilities (given as the Allan deviation) of some high-quality standards, including (from upper right to lower left) the H-maser used for our optical frequency measurements, the LPTF Cs atomic fountain operated with the UWA cryogenic sapphire oscillator [22], our atomic Ca optical standard compared to the Ca reference cavity [10], and two optical cavities used for the Hg^+ optical standard [18].

To reach a fractional frequency uncertainty of $1\text{x}10^{-18}$ in a reasonable averaging time (say one day) it will require short-term instabilities of the atomic reference of $\approx 3\text{x}10^{-16}\tau^{-1/2}$. However, if we are limited to quartz-based local oscillators we will start averaging at about $1\text{x}10^{-13}\ \tau^{-1/2}$ and will require an impractical $\tau = 10^{10}$ s (≈300 yr) to reach $1\text{x}10^{-18}$.

Since the atomic frequency instability scales as $1/\nu_0$, all else being equal, the shift from microwave to optical frequencies should improve the short-term stability by a factor of 10^5. Thus, we can imagine future optical standards using atomic-fountain methods (similar to those used in today's microwave fountain clocks) with linewidths of about 1 Hz and 10^6 atoms detected every 0.5 seconds. Theoretically, these systems could support an instability $\sigma_y(\tau) \approx 2\text{x}10^{-19}\tau^{-1/2}$. This simplistic estimate ignores significant complications that will degrade the performance. Nonetheless, it promises that in the years ahead there will be plenty of room for improvement using optical frequency standards.

IV. FEMTOSECOND OPTICAL FREQUENCY METROLOGY

The breakthrough demonstration in 1999 by the H≅nsch group [24,25] at MPQ (Max-Planck-Institut fηr Quantenoptik, Garching) showing that femtosecond mode-locked lasers can be used to span large optical frequency intervals accurately, has resulted in a total redirection of the field of optical frequency measurements. Combined with the result from JILA and MPQ, showing that nonlinear microstructure fibers can extend the frequency comb to over an optical octave of discrete resolvable lines [26,27,28], means that essentially any optical frequency can be measured with a fairly simple system. Thanks to their efforts, we now have a practical optical "clockwork" that can be used to count optical frequencies, and divide optical frequencies down to countable microwave frequencies. All the necessary ingredients are now in place for the next generation of optical frequency standards and clocks.

A. Femtosecond optical- frequency measuring system

Our version of the stabilized optical frequency comb is based on a 1 GHz repetition rate mode-locked Ti:Sapphire ring laser [29] and a nonlinear microstructure fiber [30,31]. The system is shown schematically in fig. 4, while more detail can be found in reference [32]. The laser produces ≈25 fs pulses with a spectral bandwidth of ≈30 nm (-3 dB) centered at 800 nm. When approximately 600 mW from the laser is focused into a 10 to 20 cm length of microstructure fiber (core size ≈1.7 μm) the transmitted power is about 300 mW. The fiber broadens the comb to just over an octave of spectral bandwidth, spanning from 520 nm to 1070 nm.

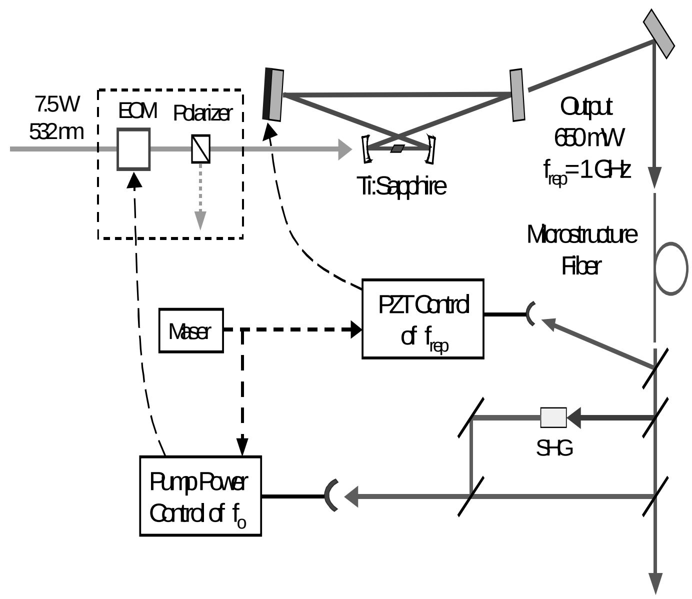

Fig. 4. Femtosecond-laser-based optical "clockwork" with a one gigahertz repetition rate. The output of the mode-locked Ti:Sapphire ring laser is broadened in a microstructure fiber. The IR portion is then frequency-doubled (SHG) back to the green using $KNbO_3$ and recombined with the original green from the fiber. This produces a signal at the offset frequency (f_0) that is locked to the H-maser via the electro-optic modulator in the pump beam. A second servo system drives the PZT on the ring cavity to control the repetition rate, f_{rep} .

The repetitive train of pulses produced by a stable mode-locked laser appears in the frequency domain as a comb of discrete modes separated in frequency by the pulse repetition rate, f_{rep} . If the evenly spaced comb of optical frequencies were extrapolated to zero frequency there would, in general, be an offset from zero by some amount f_0 . The offset, f_0 , is understood as resulting from the difference between the group and phase velocities for the ultra-short pulse traversing the laser cavity [24,26]. Thus, the resulting optical comb of modes can be described with two parameters, the spacing between the modes given by the pulse repetition rate f_{rep}, and the offset f_0. The frequency of any mode of the optical comb can be written as: $f(m) = f_0 + m \cdot f_{rep}$, where m is an integer.

The repetition rate f_{rep} is easily detected in a fast photodiode that monitors light coming out of the microstructure fiber. The offset frequency f_0 can be detected using the “self-referencing” method developed at JILA and MPQ [26,28]. This method is shown schematically in fig.4. The infrared portion of the optical comb (near 1100 nm) from the microstructure fiber is frequency doubled in a $KNbO_3$ crystal to generate light at 520 nm, which is then recombined (after an optical delay line) on a photodiode with the 520 nm light generated in the microstructure fiber. The resulting beatnote gives the desired offset frequency,

$$f_0 = 2(f_0 + m \cdot f_{rep}) - (f_0 + 2m \cdot f_{rep}) .$$

B. *Optical frequency measurement results*

Our femtosecond-laser-based optical clockwork was previously used to measure the absolute frequencies of both the Hg^+ standard and the Ca standard relative to the definition of the second as realized by the Cs primary frequency standard at NIST [19]. This was accomplished by phase-locking both the repetition rate and offset frequency of the femtosecond-comb to high-quality synthesizers referenced to a 5 MHz signal provided by a hydrogen maser that is part of the NIST time scale. With f_{rep} and f_0 thus fixed, we measure the absolute frequency of Hg^+ (563 THz, at the dye laser wavelength of 563 nm) and Ca (456 THz, 657 nm) by counting the beatnotes between the CW lasers locked to the atoms and a nearby mode of the comb. These beatnotes are detected on Si-PIN photodiodes, amplified by ≈50dB, bandpass filtered (bandwidth ≈10 MHz at 200 MHz) and counted. Typically we achieve a signal-to-noise ratio on the detected beatnotes of about 30 to 40 dB in a detection bandwidth of 300 kHz, which is usually sufficient for reliable counting. The frequencies of the optical standards can then be written as,

$$f_{opt} = m_0 \cdot f_{rep} + f_0 \pm f_{beat} .$$

The large integer $m_0 \approx 5 \times 10^5$ can be determined from knowledge of the approximate optical frequency or by making measurements with different values of f_{rep}. In our case f_{rep} is ≈1 GHz, so knowing f_{opt} with a precision of 400 MHz or better unambiguously determines m_0. An interferometric measurement with a wavelength meter is convenient for this purpose.

The results of our measurements of the Hg^+ and Ca optical frequencies relative to the Cs primary standard are [19]

$f(Hg^+) = 1,064,721,609,899,143(10)$ Hz and

$f(Ca) = 455,986,240,494,158(26)$ Hz.

Given fractionally these are,

$\Delta f(Hg^+) / f(Hg^+) \approx 9.4 x 10^{-15}$ and

$\Delta f(Ca) / f(Ca) \approx 5.7 x 10^{-14}$. The Hg^+ result represents one of the highest accuracy optical frequency measurements to date. The Ca measurement has the highest accuracy yet reported for Ca, and is in good agreement with previous measurements made at the PTB-Braunschweig using a traditional optical frequency chain [33] and is also consistent with a recent Ca measurement made using a femtosecond comb [34]. A summary of the historical record of frequency measurements of laser-cooled Ca frequency standards is shown in fig. 5.

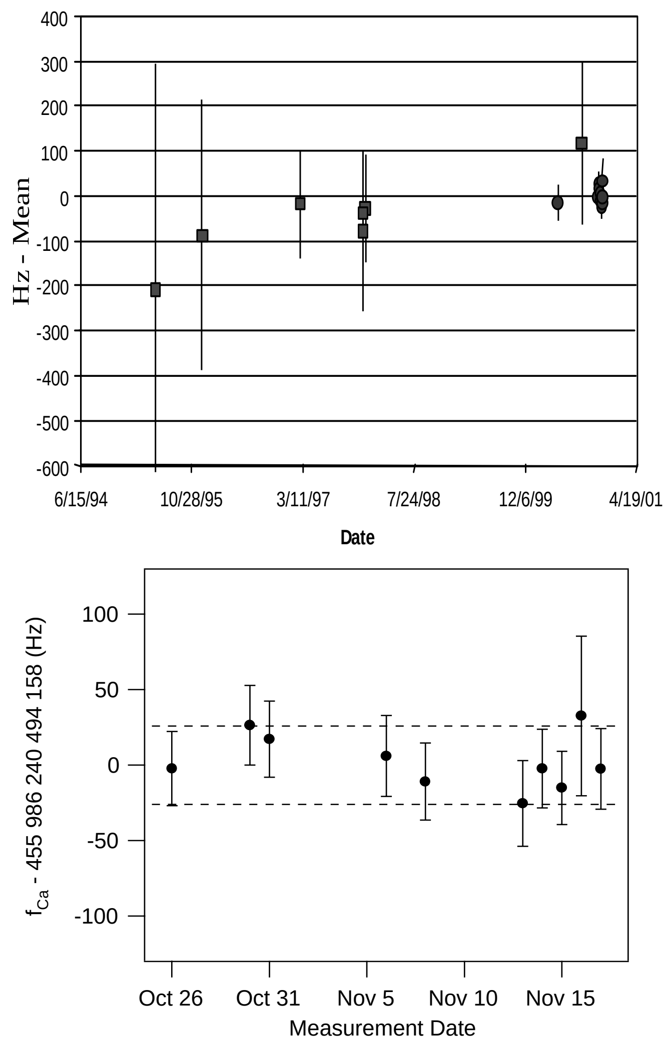

Fig. 5. Frequency measurements of laser-cooled Ca optical frequency standards over the past five years are shown in the upper half of the figure. Here the rectangular data points represent published data from the PTB [33,34], while the round data points are recently reported measurements made in our laboratory [19]. A harmonic frequency chain was used for the PTB measurements prior to 1999, while femtosecond optical combs were used by both laboratories for the measurements made in 2000. The lower half of the figure plots an expanded view of our measurements taken in Oct. and Nov. of 2000. Our data points represent daily averages of about 10 measurements, with an average duration of about 400 seconds each. On both plots the vertical axes are in hertz and are relative to the mean value of our measurements.

The consistency of the Ca results between laboratories, over time, and with different apparatus demonstrates the good reproducibility of these standards with respect to the Cs primary frequency standards.

Very recently we remeasured the Hg^+ frequency with respect to the NIST F-1 primary standard. Our new measurement is plotted in fig. 6 along with our previous results. For the new measurement the femtosecond-optical comb was configured slightly differently, but the results are in excellent agreement. In Aug. 2000 the statistical uncertainty of the measurement of the Hg^+ (1 064 THz) frequency was +/- 2.4 Hz, and for the measurement of Feb.9th, 2001, it was +/- 4.7 Hz. The measurement imprecision is determined in part by the maser instability combined with our averaging times, and in part by the accuracy determination of the Cs primary standard, which contributes about +/- 2 Hz at 1,064 THz.

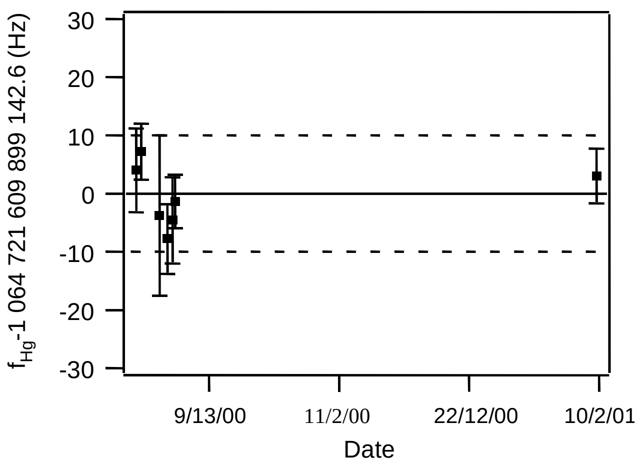

Fig. 6. Hg^+ frequency measurements verses the date. These measurements occurred in two groups, Aug. 2000 and Feb. 2001. Each data point corresponds to a daily average of the frequency measurement corresponding to typically 3000 s of total time averaging per day. The error bars indicate the statistical uncertainty of the daily measurements and the two dotted lines indicate the +/- 10 Hz uncertainty assigned to the Hg^+ standard in lieu of an accuracy evaluation.

The excellent agreement of the Hg^+ measurements over time is very encouraging. We also note that the experimental data is well clustered within the +/- 10 Hz absolute uncertainty that has been assigned based on theoretical arguments in lieu of a complete accuracy evaluation [19]. Further improvements in this absolute frequency measurement will rely on an accuracy evaluation of the Hg^+ standard and improvements in the realization of the second by the Cs primary standard. Even with the ± 10 Hz uncertainty, the agreement in the Hg^+ measurements over the time interval of 169 days provides an improved limit on any relative time variation of the Hg^+ frequency compared to the Cs definition of the second, thus $1/f_{Hg^+}(\partial f_{Hg^+}/\partial t) = (+0.6 \pm 3)x10^{-14}\ yr^{-1}$.
Similarly, our Ca measurements combined with the PTB measurements from 1997 shown in Fig. 5 give $1/f_{Ca}(\partial f_{Ca}/\partial t) = (+2 \pm 8)x10^{-14}\ yr^{-1}$, as previously reported [19]. Here t represents "time" as defined by the Cs definition of the second. We implicitly assume that any temporal variation is linear over the duration of these measurements. This type of absolute frequency intercomparison can be combined with theoretical models to put some constraints on possible time variations of fundamental constants [35,36,37,38,39].

When using the femtosecond optical comb to measure the Hg^+ and Ca lasers relative to the microwave standard, the scatter in the frequency measurements is dominated by the short-term instability of the H-maser. One of the data runs from Feb. 9th 2001 illustrates this point in Fig. 7.

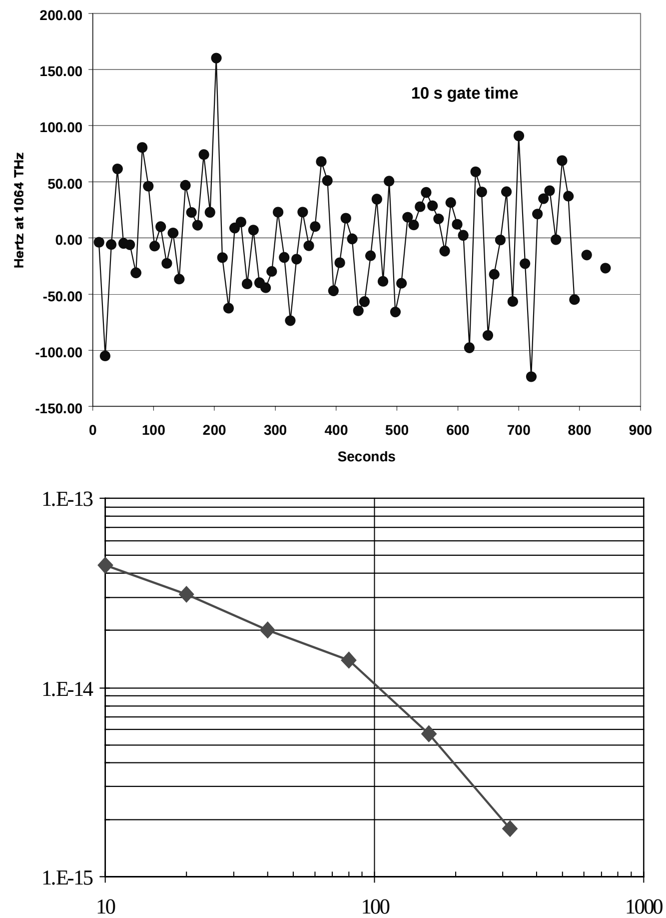

Fig. 7. The upper half of the figure shows the time-series of frequency measurements of Hg^+ in Hertz offset from the mean frequency at ≈ 1064 THz. The data was taken with a counter gate time of 10 s. From this data, (and ignoring the dead times in counting of about 150ms/pt) we calculate an approximated Allan Deviation that shows (lower part of figure) an instability consistent with that of the H-maser (within measurement uncertainties).

C. Accuracy issues and error detection

The accuracy of our optical frequency measurements comes from our knowledge of the frequency of the maser that serves as the frequency reference for the optical comb. This maser is part of the NIST time scale and is continuously monitored relative to the other components of the time scale, which consists of 5 hydrogen masers and three commercial Cs standards. Approximately once per month the time scale is recalibrated against the NIST Cs primary standard, NIST F-1, a laser cooled Cs atomic fountain with an evaluated uncertainty [40] of ≈2×10^{-15}. Used in this way the time scale provides a continuously available reference with an instability of about $2\times10^{-13}\tau^{-1/2}$ averaging down to ≈4×10^{-16} at 1 day. The absolute frequency of the maser is known with a fractional uncertainty of about ≈2×10^{-15}, as determined by the periodic comparisons of the time scale to NIST F-1.

To ensure that our measurements are not contaminated by erroneous counts or cycle-slips we include redundancy in the counting of the critical beatnotes and phase-locks as follows. After photodetection, the beatnote between the optical frequency standard and a mode of the comb is split into two paths; the primary path goes directly to the main counter. The secondary path is split into two more paths, one of which goes directly to a second counter while the other goes to a divide-by-four prescaler before going to the other input channel of the second counter. Operating the second counter in the ratio mode gives the ratio of the frequency of the directly counted beatnote to the prescaled beatnote. Since the prescalar divides by four, the output of the ratio counter should always give a result of $4.000\overline{0}$. If not, this indicates irreproducibility in counting the optical beatnote, and the results from the primary counter are then rejected. This method proves quite effective at eliminating erroneous data where the counters are not giving reproducible results. Causes of bad counts are usually related to insufficient signal size, or signal-to-noise ratio in the beatnote. The phase-locks controlling f_{rep} and f_0 of the femtosecond comb are also monitored with counters that detect phase-lock errors as described in reference [24].

The actual performance of the whole system depends on many factors; sometimes more than 50 % of the data are rejected by one of the three counters that are monitoring for counting errors. More commonly, the discarded counts represent a few percent of the data. In normal operation as a tool for measuring optical frequencies, all the synthesizers, counters and digital phase locks are referenced to a single 5 MHz signal from a hydrogen maser. The performance of the synthesizer that is used to control (or as a reference to measure) the repetition rate is critically important because its frequency is multiplied by the large integer m_0 in determining the measured optical frequency. For this synthesizer we need the highest accuracy, lowest phase-noise, and lowest phase drift that is possible. Though probably not optimal, we are currently using an HP8662A for this purpose [41]. The scatter of the optical frequency measurements (eg. $\pm$ 50 Hz for a gate time of 10 s, fig. 7) corresponds well with the instability of the maser multiplied up to the optical frequency at 1 064 THz. This indicates that, at least at this level, the synthesizer does not further degrade the short-term stability. Not surprisingly, we have observed that changes in temperature of the RF and microwave components do produce time dependent phase shifts, which appear as small frequency shifts at the optical frequency. For the critical synthesizer we measure a temperature coefficient of fractional frequency change of ≈1×10^{-14} for a temperature change of one kelvin per hour.

V. NOISE IN FEMTOSECOND OPTICAL COMBS

The simple model that we use for the frequency spectrum of the femtosecond-optical-comb, $f(m) = f_0 + m\cdot f_{rep}$, has been verified, at least in the time-averaged counting of optical beatnotes, by Holzwarth et al. [28]. By comparing two different optical synthesizers based on the same microwave reference they showed that the synthesis process gave the same optical frequency within a measurement uncertainty of ≈5×10^{-16}. The present reproducibility of our measurements of Hg^+ ($\pm$3 Hz at 10^{15} Hz) also provides additional assurance of the reproducibility of optical frequency combs in measuring absolute optical frequencies relative to the Cs primary standard.

However, these femtosecond measurement systems are built around counting optical beatnotes, and the counting process gives only the average frequency during the gate interval of the counter. It provides very little information about the fluctuations and noise properties of the signals. As we strive toward the goals of 1×10^{-18} fractional frequency uncertainty and $1\times10^{-16}\tau^{-1/2}$ instability, we will require detailed knowledge and control of the noise properties of all of the signals. We are just beginning to address these issues, some of which are fundamental (such as shot noise and thermal noise) while others are challenging technical limitations.

The femtosecond-optical comb can be configured in many different ways, but typically electronic servo systems are used to control two of the three signals (f_{beat}, f_{rep} and f_0) while the third "unknown" is then measured (counted). In the most obvious implementations we face a number of technical challenges that include: non-orthogonality in detecting the signals and the servo systems that control them [42,43], photodiode limitations, fiber noise, phase-noise in amplifiers and components (filters, cables, isolators etc. are all non-negligible), combined with temperature and mechanical instabilities.

We choose to focus here, and in the laboratory, on three issues that appear to cause the most critical and challenging limits to the development of these femtosecond mode-locked lasers for optical-frequency metrology and synthesis. With our present systems these issues are: excess amplitude noise generated by the nonlinear micro-structure fibers, residual phase-noise in f_{rep}, and limits to the photocurrent and bandwidth of the photodiode that is used to detect f_{rep}.

A. *Fiber AM noise*

As others have, we observe significant amounts of broadband AM noise that appears on the light out of the microstructure fiber when the femtosecond pulse energy is increased. The physical origin of the noise has not been studied in detail for microstructure fibers, but similar effects have been observed in telecom fibers [44]. With the microstructure fiber we observe that the magnitude of the excess AM noise has a threshold-like behavior that grows rapidly for pulse energies above ⊔ 300 pJ (pulse width ≈ 30 fs) as shown in Fig. 8.

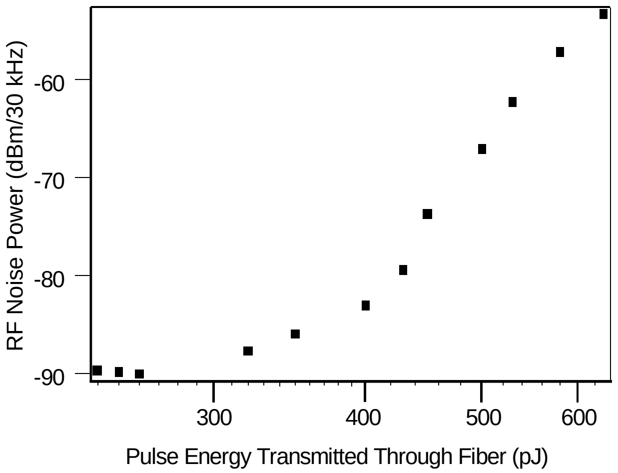

Fig. 8. Amplitude noise measured on the light (in a ≈ 2 nm optical bandwidth near 1064 nm) transmitted through the microstructure fiber is plotted as a function of the transmitted pulse energy. This data was taken using a 100 MHz repetition rate mode-locked Ti:Sapphire laser with input pulse durations of ≈ 30 fs. The vertical axis gives the average noise power in a 30 kHz bandwidth centered near 550 MHz.

Since we typically require approximately 300 pJ pulses to obtain an optical-octave of spectral broadening, the AM noise from the fiber can limit the signal-to-noise ratios achievable on the detected beatnotes. This noise appears on the full white-light spectrum as well as on spectrally resolved regions such as that shown in fig.8. Within the bandwidth of our photodetectors (1 MHz to 8 GHz) the excess noise is approximately spectrally white and creates a broad noise background under the coherent signals. The limitations attributed to fiber AM noise appear to be more serious for lasers with lower repetition rates. This is because the noise increases with pulse energy, while the average power is proportional to the pulse energy times the repetition rate. When the excess noise limits the useful optical power that we can send through the nonlinear fiber a higher repetition rate gives both a higher average power and fewer modes in the frequency comb. [32].

B. f_{rep} Phase- noise

The noise characteristics of f_{rep} are particularly critical since the connection between f_{rep} and the optical frequency is through the large integer m. When the femtosecond optical frequency comb is configured (as in fig. 4) with f_{rep} and f_0 locked to the maser, then phase-noise in the maser is multiplied up to the optical frequency. If the system were to be operated as an optical clock, with the femtosecond laser locked to the optical beatnote and f_0 locked in the usual way, then the output comes from the repetition rate, f_{rep}. Assuming that the optical frequency standard is perfectly stable but the femtosecond laser has some intrinsic noise, the servo systems have to lock to the optical beatnote signal and suppress the noise of the femtosecond laser. In this regard it is interesting to explore the intrinsic noise characteristics of the femtosecond optical comb under free-running conditions. With this information in hand, we will know what is required of the servo systems to suppress the noise sufficiently to reach a desired stability. We have measured the noise properties of the detected f_{rep} signal for a Kerr lens mode-locked Ti:sapphire laser operating with a repetition rate of ≈100 MHz. The most interesting results are shown in Fig 9.

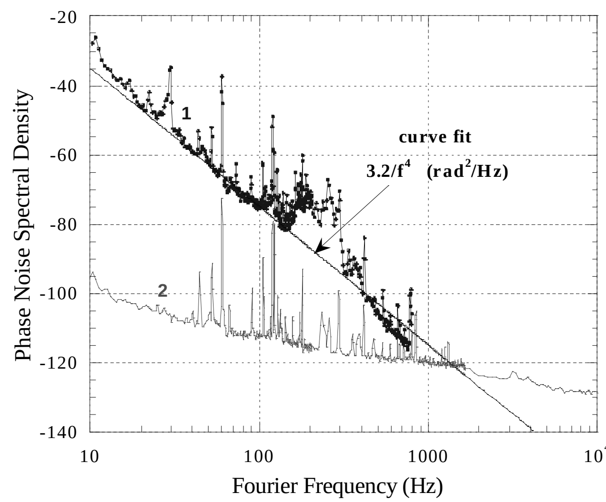

Fig. 9. Measured phase-noise spectral density of f_{rep} for a free-running Kerr-lens mode-locked Ti:sapphire laser (curve "1"). This data was taken at 900 MHz, the ninth harmonic of the laser's repetition rate. For comparison, curve "2" shows the measured phase-noise spectral density of the 8662A frequency synthesizer.

For the free-running laser, the phase-noise on f_{rep} is large at low frequencies and drops rapidly toward higher frequencies as $\approx 3.2/f^4$ (rad^2/Hz), as indicated by the solid line in fig.9. The experimental data is the measured phase noise at 900 MHz derived from the 9th harmonic of the 100 MHz repetition rate. An f^{-4} dependence of the phase-noise spectral density implies a random-walk FM process in the laser's repetition rate. It is also noteworthy that the laser's phase noise is actually very low at higher Fourier frequencies, dropping below that of a high-quality synthesizer for frequencies > 1 kHz.

The phase-noise measurements were made by comparing f_{rep} to a stable (900 MHz) signal from an HP8662A synthesizer referenced to the H-maser [41]. We employed a slow phase-lock of f_{rep} to a second HP8662A to remove low-frequency drifts (addition technical details are deferred to a subsequent publication, but some are provided in ref. [42]). We also observe that the phase-noise spectral density of f_{rep} is not identical before and after the nonlinear fiber, indicating that some additional phase-noise is by the nonlinear fiber itself. To avoid potential errors that this might cause we generally measure all signals for the servos and counting after the nonlinear fiber.

C. f_{rep} *Shot-noise*

Ultimately we want to operate these standards as optical clocks locked to the optical transitions, with the output coming as a stable microwave signal [45]. For these systems to run as optical clocks, the femtosecond optical clockwork needs to be run in the opposite direction from that indicated in fig. 4. In this case, the optical clock will provide stable frequencies out at the repetition rate and its harmonic nf_{rep}. These can be used to count time intervals, compare with other frequency standards, and can serve as a scale of time and frequency.

Ignoring for now the excess AM noise from the fiber and the other issues mentioned above, we still expect that the shot noise in the detected photocurrent will provide a fundamental limit to our ability to extract f_{rep} with high precision [42,46]. Even though the photocurrent is generated in the detector as very short pulses, we measure (directly from the laser) a noise background consistent with that calculated for the shot noise of the average detected photocurrent. The large peak power in the optical pulses (particularly in 100 MHz system) can cause distortion of the electrical waveform and spectrum even with fairly low average power on the photo-detector. It is thus challenging, but possible, to measure the shot noise directly on the photocurrent. However, it can be precisely measured with carrier-suppression methods and a two-channel measurement system [47,48].

We can estimate the stability in f_{rep} that should be achievable assuming sufficient photocurrent to be above the thermal and amplifier noise contributions. We expect a spectrally white background noise due to the shot noise, with a current noise spectral density (in $A/\sqrt{Hz}$) of $i_{sn} = \sqrt{2ei_{avg}}$ for an average detected current i_{avg}. This noise adds white phase-noise to the signal current i_n at the nth harmonic of f_{rep}. The fractional frequency instability due to white phase-noise in an averaging time τ, expressed as the Allan deviation is calculated as [46,49,50].

$$\sigma_y(\tau) = \frac{\sqrt{3S_\phi^{white}\Delta f}}{2\pi\tau n f_{rep}}.$$

Here, for a signal power P_{signal},

$$S_\phi^{white} = \frac{2\delta P_n^{PM}}{P_{signal}} = \frac{2(i_{sn}^2 R/2)}{P_{signal}}$$

is the spectral density of phase-noise due to two symmetric noise sidebands with power density δP_n^{PM}, and R is the load impedance. Combining these results gives,

$$\sigma_y^{shot}(\tau) \approx \frac{1}{2\pi n f_{rep}\tau}\sqrt{\frac{6ei_{avg}R\Delta f}{P_{signal}}}.$$

Using our present experimental numbers we garner some appreciation of the seriousness of even the ideal shot-noise-limited performance. As an example, we consider our femtosecond laser with a 1 GHz repetition rate and an average detected photocurrent of ≈4 mA. The photocurrent produces a microwave signal power of –10dBm in 50 Ohms at 3 GHz (the 3rd harmonic). With a filter bandwidth of 5 % (150 MHz at 3 GHz) the equation above predicts a shot-noise-limited instability of $\sigma_y^{shot}(\tau) \approx 3x10^{-14}\tau^{-1}$. Possible methods to improve this stability include: detecting multiple harmonics, using a very high harmonic n, using a high-Q resonator [51] to limit the bandwidth Δf, and measuring pulse timing rather than RF phase. Just narrowing the filter bandwidth from 150 to 1 MHz will improve the predicted shot-noise-limited performance to a more acceptable $\approx 2x10^{-15}\tau^{-1}$. In any case, because of the shot-noise limitation, generating microwave signals with high spectral purity from the repetition rate will require high-speed photodetectors that can handle high power and that have reasonable reponsivity over the spectral range from 500 to 1000 nm.

VI. SUMMARY

After many years of development and continued progress in high-resolution spectroscopy, stable lasers, and atom cooling and trapping, the self-referenced optical-frequency comb provides the missing link to the realization of the next generation of frequency standards and clocks. The new methods of femtosecond optical frequency metrology seem well suited for measuring the absolute frequency of essentially any optical frequency standard with a reproducibility, as demonstrated here, at the level of the present primary atomic frequency standards. There are however, some serious fundamental and technical challenges that must be addressed if we hope to realize the next factor of 1000 improvement that appears to be possible.

The frequencies of our Hg^+ and Ca optical standards were measured using a 1 GHz optical frequency comb and the resulting values are in good agreement with previous measurements. In the case of the Hg^+ standard, the demonstrated reproducibility is at least as good as the present ability to measure absolute frequency, which in fractional frequency is $\approx 3x10^{-15}$. This is limited in part by the short-term instability of the maser combined with the limited duration of our measurements, and largely by the uncertainty of the primary Cs atomic fountain, $\approx 2x10^{-15}$.

Better short-term stability is required to make phase, frequency and timing measurements quickly and to improve the accuracy of frequency standards. Both the Ca and Hg^+ standards have already demonstrated short-term instability in the 10^{-15} range for averaging times of 1 second and longer. The microwave atomic standards do not have adequate stability to fully test the optical standards, so progress on this front will likely come from direct intercomparisons between optical frequency standards.

The tremendous progress in this field during the past two years brings us to the point that the optical standards are now pushing hard against the performance limitations of the primary microwave standards. Furthermore, we see no evidence that the rate of improvement in optical frequency standards and measurement systems will be slowing in the near future. As more optical standards come into operation, direct intercomparisons should allow us to take advantage of the excellent short-term stability to explore into the 10^{-17} range of stability and accuracy with reasonably short averaging times. In the longer term, the improved accuracy and stability of optical frequency standards will have applications in navigation and communication systems. Following as a natural byproduct will be improved knowledge of the fundamental constants, atomic structure, more powerful and compelling searches for time variation of fundamental "constants", tests of our theoretical model of space-time, and fundamental symmetries of nature.

VII. Appendix A: Atomic Stability Analysis

We estimate the fractional frequency instability of an atomic frequency standard at center frequency ν_0, assuming: atom projection noise limited detection, no local oscillator noise, and Ramsey separated fields excitation. For now, we also assume Ramsey fringes with 100% contrast. The fluctuation in the number of atoms that have made a transition (atom projection-noise) is

$$\Delta N_0 = \sqrt{N_0 p(1-p)}$$

where p = transition probability. The signal is given by

$$Signal = n_{ph} p N_0 \quad ,$$

where n_{ph} is the mean number of scattered photons per atom that are detected in a single measurement. The total noise from the atom projection and photon shot noises is

$$N_{tot} = n_{ph}\sqrt{N_0 p(1-p+\frac{1}{n_{ph}})} \quad .$$

In the atom projection-noise limit we drop the second term as small compared to the first, and the total noise from one side of the atomic resonance becomes

$$N_{tot} = n_{ph}\sqrt{N_0 p(1-p)} \quad .$$

Measuring on one side of the fringe at a detuning from resonance δ for a Ramsey time T_R produces a signal

$$S = N_0 n_{ph}(1+cos[2\pi\delta T_R])/2 \quad .$$

Now taking

$$\delta = \delta_0 \pm \varepsilon, \; and \; \delta_0 = 1/4T_R$$

where ε is the error in frequency tuning, we obtain signals S_+ and S_- from opposite sides of the fringe,

$$S_{+,-} = N_0 n_{ph}(1 \pm sin[2\pi\varepsilon T_R])/2 \quad .$$

The servo signal, $\frac{\partial(S_+ - S_-)}{\partial\varepsilon}$, is derived from a measurement of both sides of the fringe, which is just the difference between S_+ and S_-

$$S_+ - S_- = n_{ph} N_0 sin[2\pi\varepsilon T_R]$$

$$\frac{\partial(S_+ - S_-)}{\partial\varepsilon} = 2\pi T_R n_{ph} N_0 cos[2\pi T_R \varepsilon] \approx 2\pi T_R n_{ph} N_0$$

Noise NN, upon processing the signal, is then

$$NN = \sqrt{N_{tot+}^{\;2} + N_{tot-}^{\;2}} = n_{ph}\sqrt{N_0[p_+(1-p_+) + p_-(1-p_-)]} \quad .$$

The RMS frequency error for a measurement of both sides of the fringe can then be written as

$$\delta\nu_{rms} = \frac{NN}{\frac{\partial(S_+ - S_-)}{\partial\varepsilon}} = \frac{\sqrt{N_0[p_+(1-p_+) + p_-(1-p_-)]}}{2\pi T_R N_0} \quad .$$

Taking $\Delta\nu_{Ram} = \frac{1}{2T_R}$ and $p_+ \approx p_- \approx \frac{1}{2}$ we obtain $\delta\nu_{rms}$ which represents the RMS frequency error for a two-sided measurement, where the averaging time τ, is the same as the cycle time T_c, and N_0 is the number of atoms detected on one side of the atomic resonance,

$$\delta\nu_{rms} = \frac{\Delta\nu_{Ram}}{\pi}\frac{1}{\sqrt{2N_0}} \quad .$$

For a measurement time τ that is longer than the cycle time T_c, the total number of atoms detected in the measurement is larger than N_0 by the ratio τ/T_c, thus we have

$$\delta\nu(\tau)_{rms} = \frac{\Delta\nu_{Ram}}{\pi}\sqrt{\frac{T_C}{2N_0\tau}} \quad .$$

Finally, we can form the expression for stability as given by the Allan deviation,

$$\sigma_y(\tau) = \frac{\delta\nu(\tau)_{rms}}{\nu_0} = \frac{\Delta\nu_{Ram}}{\pi\nu_0}\sqrt{\frac{T_C}{2N_0\tau}} \quad .$$

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[1] See invited papers by J.L. Hall and by T.W. H≅nsch in this issue.

[2] D.A. Jennings, C.R. Pollock, F.R. Petersen, R.E. Drullinger, K.M. Evenson, J.S. Wells, J.L. Hall, and H.P.Layer, "Direct frequency measurement of the I_2-stabilized He-Ne 473 THz (633 nm) laser", Optics Lett. **8**, 136-138 (1983).

[3] D.A. Jennings, K.M. Evenson, and D. J.E. Knight, "Optical Frequency Measurements", Proc. IEEE, **74**, 168-179 (1986). J.L. Hall, "Optical Frequency Measurement: 40 Years of Technology Revolutions," IEEE J. Selected Topics in Quant. Electron., **6**, pp. 1136-1144 (2000).

[4] J.C. Bergquist, W.M. Itano, and D.J. Wineland, "Laser Stabilization to a Single Ion", in *Frontiers in Laser Spectroscopy, proceedings of the international school of physics "Enrico Fermi", course 120,* ed. By T.W. H≅nsch and M. Inguscio (North-Holland, Amsterdam, pp. 359-376 (1994). J.C. Bergquist, R.J. Rafac, B.C. Young, J.A. Beall, W. M. Itano and D.J. Wineland, "Sub-dekaHertz, Spectroscopy of $^{199}Hg^+$," Proc. SPIE vol. **4269,** *Laser Frequency Stabilization Standards, Measurement and Applications*, San Jose, CA, pp. 1-7 (2001).

[5] C.W. Oates, F. Bondu, R.W. Fox and L. Hollberg, "A Diode-Laser Optical Frequency Standard Based on Laser-Cooled Ca Atoms: Sub-kilohertz Spectroscopy by Optical Shelving Detection, Eur. Phys. J. **D7**, 449-460 (1999).

[6] H.G. Dehmelt, "Mono-Ion Oscillator as Potential Ultimate Laser Frequency Standard," IEEE Trans. Instrum. Meas. **IM-31**, 83-87 (1982).

[7] D.J. Wineland, J.C. Bergquist, J.J. Bollinger, W.M. Itano, D.J. Heinzen, S.L. Gilbert, C.H. Manney, and M.G. Raizen, "Progress at NIST Toward Absolute Frequency Standards Using Stored Ions," IEEE Trans. Ultrason. Ferroelect. and Freq. Control, 37, pp. 515-523 (1990).

[8] A.A. Madej and J.E. Bernard, "Single-Ion Optical Frequency Standards and Measurement of their Absolute Optical frequency," in *Frequency Measurement and Control*, Topics Appl. Phys. **79**, A. N. Luiten (Ed.) (Springer-Verlag, Berlin) pp. 153-194 (2001).

[9] P.T.H. Fisk, "Trapped-Ion and Trapped-Atom Microwave Frequency Standards," Rep. Prog. Phys., **60**, 761-817 (1997).

[10] B. C. Young, F. C. Cruz, W. M. Itano, and J. C. Bergquist, "Visible Lasers with SubHertz Linewidths", Phys. Rev. Lett., **82**, pp. 3799-3802 (1999).

[11] R. J. Rafac, B. C. Young, J. A. Beall, W. M. Itano, D. J. Wineland, and J. C. Bergquist, "Sub-dekahertz Ultraviolet Spectroscopy of 199Hg+", Phys. Rev. Lett., **85**, pp. 2462-2465 (2000).

[12] D.J. Wineland, J.C. Bergquist, W.M. Itano, F. Diedrich, and C.S. Weimer, "Frequency standards in the optical spectrum," in *The Hydrogen Atom*, edited by G.F. Bassani, M. Inguscio, and T.W. H≅nsch (Springer-Verlag, Berlin) 123-133 (1989).

[13] J.L. Hall, M. Zhu and P. Buch, "Prospects for Using Laser-Prepared Atomic Fountains for Optical Frequency Standards Applications," J. Opt. Soc. Am B, **6**, pp. 2194-2205 (1989).

[14] Ye.V. Baklanov, and B.Ya. Dubetsky, and V.P. Chebotayev, "Non-Linear Ramsey Resonance in the Optical Region" Appl. Phys. **9**, 171-173 (1976).

[15] J.C. Bergquist, S.A. Lee, and J.L. Hall, "Saturated Absorption with Spatially Separated Laser Fields: Observation of Optical Ramsey Fringes," Phys. Rev. Lett., **38**, pp. 159-162 (1977).

[16] Ch. J. Bordϑ, Ch. Salomon, S. Avrillier, A. Van Lerberghe, Ch. Brϑant, D. Bassi, G. Scoles, "Optical Ramsey Fringes with Traveling Waves," Phys. Rev. A, **30**, pp. 1836-1848 (1984).

[17] U. Sterr, K. Sengstock, W. Ertmer, F. Riehle, J. Helmcke, "Atom Interferometry Based on Separated Light Fields", in *Atom Interferometry*, edit. P. Berman, Academic Press, pp. 293-362 (1997).

[18] C. W. Oates, E. A. Curtis, and L. Hollberg, "Improved short-term stability of optical frequency standards: approaching 1 Hz in 1s with the Ca standard at 657 nm," Opt. Lett., **25**, pp. 1603-1605 (2000).

[19] Th. Udem, S. A. Diddams, K. R. Vogel, C. W. Oates, E. A. Curtis, W. D. Lee, W. M. Itano, R. E. Drullinger, J. C. Bergquist, and L. Hollberg, "Absolute Frequency Measurements of the Hg+ and Ca Optical Clock Transitions with a Femtosecond Laser," Phys. Rev. Lett., **86**, pp. 4996-4999 (2001).

[20] This is a reasonable assumption based on results of reference [7]. We note that excellent short-term instabilities have also been achieved with microwave oscillators using cyrogenic sapphire resonators: A. Luiten, A. Mann, M. Costa, and D. Blair, IEEE Trans. Instrum. Meas. **44**, 132 (1995). S. Chang, A.G. Mann and A.N. Luiten, "Cryogenic Sapphire Oscillator with Improved Frequency Stability," Proc. IEEE/EIA – International Freq. Control Symposium, pp 475-479 (2000). G.J. Dick and R.T. Wang "Stability and Phase Noise Tests of Two Cryo-cooled Sapphire Oscillators," Proc. EFTF – IEEE International Freq. Control Symposium, pp. 548-551 (1999).

[21] W. M. Itano, J.C. Bergquist, J.J. Bollinger, J.M. Gilligan, D.J. Heinzen, F.L. Moore, M.G. Raizen, and D.J. Wineland, "Quantum Projection Noise: Population Fluctuations in Two-Level Systems", Phys. Rev. A, **47**, 3554-3570 (1993).

[22] G. Santarelli, Ph. Laurent, P. Lemonde, A. Clairon, A.G. Mann, S Chang, and A. N. Luiten, "Quantum Projection Noise in an Atomic Fountain: A High Stability Cesium Frequency Standard", Phys. Rev. Lett., **82**, 4619-4622 (1999).

[23] C. Fertig and K. Gibble, "Measurement and Cancellation of the Cold Collision Frequency Shift in an ^{87}Rb Fountain Clock", Phys. Rev. Lett., **85**, 1622-1625 (2000). Y. Sortais, S. Bize, C. Nicolas, G. Santarelli, C. Salomon, A. Clairon, "^{87}Rb Versus ^{133}Cs in Cold Atom

Fountains: A Comparison," IEEE Trans. Ultrason. Ferroelect. and Freq. Control, **47**, 1093-1097 (2000).
[24] Th. Udem, J. Reichert, R. Holzwarth, and T.W. Hänsch, "Accurate Measurement of Large Optical Frequency Differences with a Mode-Locked Laser," Optics Lett.,**24**, pp. 881-883 (1999).
[25] J. Reichert, R. Holzwarth, Th. Udem, and T. W. Hänsch, "Measuring the frequency of light with mode-locked lasers," Opt. Comm., **172**, pp. 59-68 (1999).
[26] D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, "Carrier-envelope phase contol of femtosecond mode-locked lasers and direct optical frequency synthesis," Science, **288**, pp. 635-639 (2000).
[27] S.A. Diddams, D.J. Jones, J. Ye, S.T. Cundiff, J.L. Hall, J.K. Ranka, R.S. Windeler, R. Holzwarth, T. Udem and T.W. Hänsch, "Direct Link between Microwave and Optical Frequencies with a 300 THz Femtosecond Laser Comb," Phys. Rev. Lett., **84**, pp. 5102-5105 (2000).
[28] R. Holzwarth, Th. Udem, T. W. Hänsch, J. C. Knight, W. J. Wadsworth, and P. St. J. Russell, "Optical Frequency Synthesizer for Precision Spectroscopy," Phys. Rev. Lett., **85**, pp. 2264-2267 (2000).
[29] A. Bartels, T. Dekorsky, and H. Kurz, "Femtosecond Ti:sapphire ring laser with a 2-GHz repetition rate and its application in time-resolved spectroscopy," Opt. Lett., **24**, pp. 996-998 (1999).
[30] J. K. Ranka, R. S. Windeler, and A. J. Stentz, "Visible Continuum Generation in Air-Silica Microstructure Optical Fibers with Anomalous Dispersion at 800 nm," *Opt. Lett.*, **25**, pp. 25-27 (2000); J. K. Ranka, R. S. Windeler, and A. J. Stentz, "Optical properties of high-delta air-silica microstructure optical fibers," Opt. Lett., **25**, pp. 796-798, (2000).
[31] W. J. Wadsworth, J. C Knight, A. Ortigosa-Blanch, J. Arriaga, E. Silvestre, and P. St. J. Russell, "Soliton effects in photonic crystal fibers at 850 nm," Electron. Lett., **36**, pp. 53-54 (2000).
[32] S. A. Diddams, Th. Udem, K. R. Vogel, C. W. Oates, E. A. Curtis, R. S. Windeler, A. Bartels, J. C. Bergquist, and L. Hollberg, , "A Compact Femtosecond-Laser-Based Optical Clockwork", Proc. SPIE vol. **4269,** *Laser Frequency Stabilization Standards, Measurement and Applications*, San Jose, CA, pp.77-83 (2001).
[33] H. Schnatz, B. Lipphardt, J. Helmcke, F. Riehle, and G. Zinner, "First Phase-Coherent Frequency Measurement of Visible Radiation," Phys. Rev. Lett., **76**, pp. 18-21 (1996). J F. Riehle, H. Schnatz B. Lipphardt, G. Zinner, T. Trebst and J. Helmcke, "The Optical Calcium Freqeuncy Standard", IEEE Trans. Instrum. Meas., **48**, 613-617 (1999).
[34] J. Stenger, T. Binnewies, G. Wilpers, F. Riehle, H. R. Telle, J. K Ranka, R. S. Windeler, and A. J. Stentz, "Phase-Coherent Frequency Measurement of the Ca Intercombination Line at 657 nm with a Kerr-Lens Mode-Locked Femtosecond Laser," Phys. Rev. A, **63**, 021802(R) (2001).
[35] J.D. Prestage, R.L. Tjoelker,and L. Maleki, "Atomic Clocks and Variations of the Fine Structure Constant", Phys. Rev. Lett., **18**, 3511-3514 (1995).
[36] S.V. Karsenboim, "Some Possibilities for Laboratory Searches for Variations of the Fundamental Constants", Can. J. Phys., **78**, 639-678 (2000).
[37] J.K. Webb, V.V. Flambaum, C.W. Churchill, M.J. Drinkwater, and J.D. Barrow, "Search for Time Variation of the Fine Structure Constant", Phys. Rev. Lett., **82**, pp. 884-887 (1999).
[38] V. Kosteleckϖ, editor, *Meeting on CPT and Lorentz Symmetry*, World Scientific (1999).
[39] D. Bear, R.E. Stoner, R.L. Walsworth, V.A. Kosteleckϖ and C.D. Lane, "Limit on Lorentz and CPT Violation of the Neutron Using a Two-Species Nobel-Gas Maser", Phys. Rev. Lett., **85**, pp. 5038-5041 (2000).
[40] S.R. Jefferts, D.M. Meekhof, J.H. Shirley, M Stepanovic, and T.E. Parker, "Accuracy Results from NIST-F1 Laser-Cooled Cesium Primary Frequency Standard", Proc. IEEE-International. Freq. Control. Symposium, pp. 714-717 (2000).
[41] Mention of a specific product is for technical clarity only and does not represent a recommendation, other products may be better suited for this application.
[42] E. Ivanov, S. A. Diddams, and L. Hollberg, "Experimental study of the noise properties of a Ti:sapphire femtosecond laser," Proc. IEEE-International. Freq. Control. Symposium (2001).
[43] J.Ye, J.L. Hall, and S.A. Diddams, "Precisiion Phase Control of an Ultrawide-Bandwidth Femtosecond Laser: A Network of Ultrastable Frequency Marks Across the Visible Spectrum," Optics Lett. **25**, 1675-1677 (2000).
[44] M. Nakazawa, K. Tamura, H. Kubota, and E. Yoshisda, "Coherence Degradation in the Process of Supercontinuum Generation in an Optical Fiber," Optical Fiber Technology, **4**, 215-223 (1998).
[45] S.A. Diddams, Th. Udem, J.C. Bergquist, E.A. Curtis, R.E. Drullinger, L. Hollberg, W.M. Itano, W.D. Lee, C.W. Oates, K.R. Vogel, and D.J. Wineland, "An Optical clock Based on a Single Trapped $^{199}Hg^{+}$ Ion", to be published Science, July (2001).
[46] F. Walls, "Phase Noise Issues in Femtosecond Lasers" SPIE vol. **4269,** *Laser Frequency Stabilization, Standards, Measurement and Applications*, San Jose, CA, pp. 170-177 (2001).
[47] E. N. Ivanov, M. E. Tobar and R. A. Woode, "Microwave Interferometry: Application to Precision Measurements and Noise Reduction Techniques, IEEE Trans. Ultrason. Ferroelect. and Freq. Control, pp. 1526-1536 (1997).
[48] E. Rubiola, V. Giordano and J. Groslambert, "Double Correlating Interferometer Scheme for Measuring PM and AM Noise", Electron. Lett., vol. **34**, no. 1, pp. 93-94 (1998).
[49] J.A. Barnes, A. R. Chi, L. S. Cutler, D. J. Healey, D.B. Leeson, T. E. McGunigal, J. A. Mullen Jr., W. L. Smith, R. L. Sydnor, R. F. C. Vessot, and G. M. R. Winkler,

"Characterization of Frequency Stablity," IEEE Trans. Instrum. and Meas, **IM-20**, pp. 105-120.
[50] J. Rutman, "Characterization of Phase and Frequency Instabilities in Precision Frequency Sources: Fifteen Years of Progress," Proc. IEEE, **66**, 1048-1075 (1978).
[51] E.N. Ivanov, D.G. Blair and V.I. Kalinichev, "Approximate approach to the design of shielded dielectric disk resonators with whispering-gallery modes," IEEE-MTT, **41**, N 4, pp. 632-638 (1993).